
Decontamination of Radionuclides from Concrete During and After Thermal Treatment

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Research Objective

This project will determine the effect of heating concrete on its engineering properties and the potential to remove common DOE radionuclide contaminants including ^{137}Cs , ^{90}Sr , ^{60}Co , and U. In the chemical properties subtask, effects on direct volatilization of radionuclides during heating, as well as their subsequent extractability behavior, will be established over a range of temperatures up to the melting point of Portland cement. The main objective of the mechanics and thermodynamics subtask of this investigation is to develop a powerful computational capability to evaluate the effects of various regimes of rapid heating of a contaminated concrete wall or slab.

Research Progress and Implications

This report summarizes research progress after 8 months (through June 1, 1999) of a 3-year project. In the laboratory studies of radioisotopic behavior, techniques for spiking Portland cement and following volatility of four radionuclides (^{134}Cs , ^{85}Sr , ^{57}Co , and natural U) have been completed. A sequential extraction procedure for Portland cement has been adapted to assay these radionuclides and their behavior in unheated cement determined as a baseline to compare responses after thermal treatments. A large uniform batch of hydrated (cured) Portland cement was prepared and pulverized to a narrow range of particle size (-100/+200 mesh) to provide a high-surface area/unit weight of cement and will facilitate following the volatile behavior of hundreds of radioisotopically-spiked cement samples and thousands of extractions of thermally perturbed cement samples. One gram samples of this pulverized cement, contained in 2-mL capacity refractory (alumina) crucibles, are spiked with the desired radioisotope and, after aging for several weeks, exposed to various temperatures and/or heating regimes. Retention of radioisotopes is assayed non-destructively, using gamma-ray spectroscopy of the crucibles, before and after thermal treatment. Preliminary results of the retention of these four radioisotopes by cement when heated up to 1400°C (cement melted) are presented in Figure 1 along with retention of moisture, calculated from the decreasing weight. Complete volatility of ^{134}Cs was observed at temperatures of 1200°C or greater. Thus, decontamination of ^{137}Cs from cement should prove quite feasible by direct heating. However, only minor volatility of the other radionuclides was observed so that their potential for decontamination will likely depend on their modified extraction behavior in thermally-perturbed cement.

The sequential extraction of the four radionuclides has only been completed for the thermally-unperturbed spiked cement samples (Figure 2). Both ^{134}Cs and ^{85}Sr were largely removed from cement in the first stages of the sequential extractions when only water was employed. This should be contrasted to their expected behavior if they had been incorporated into the cement during or prior to curing where they would be much less extractable because they would be incorporated within the hydrated silicate minerals rather than mostly adsorbed on surfaces. The highly alkaline nature of Portland cement, with its large excess $\text{Ca}(\text{OH})_2$, probably caused the delay in extraction of both ^{57}Co and U until dilute hydrochloric acid was employed because these elements and radionuclides form highly insoluble hydroxides. Indeed, the release of both these radionuclides was delayed until the pH of the extracts decreased sharply, presumably after the excess $\text{Ca}(\text{OH})_2$ had been neutralized by

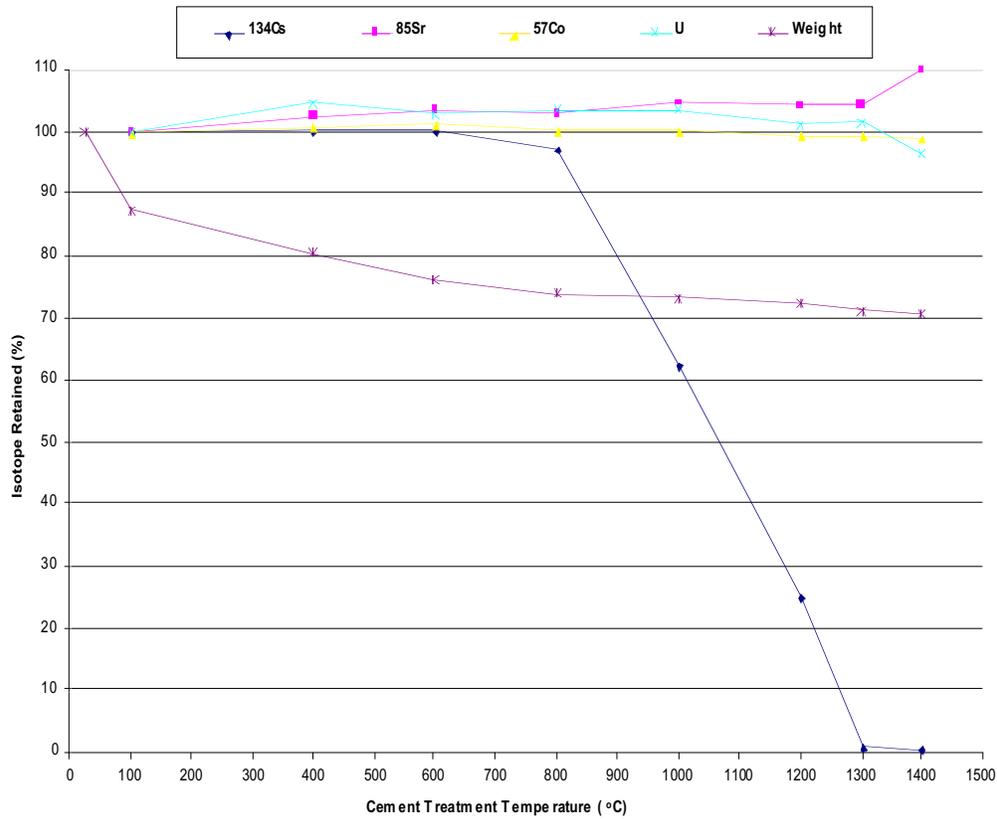


Figure 1. Retention of radionuclides by Portland cement after heating to various temperatures.

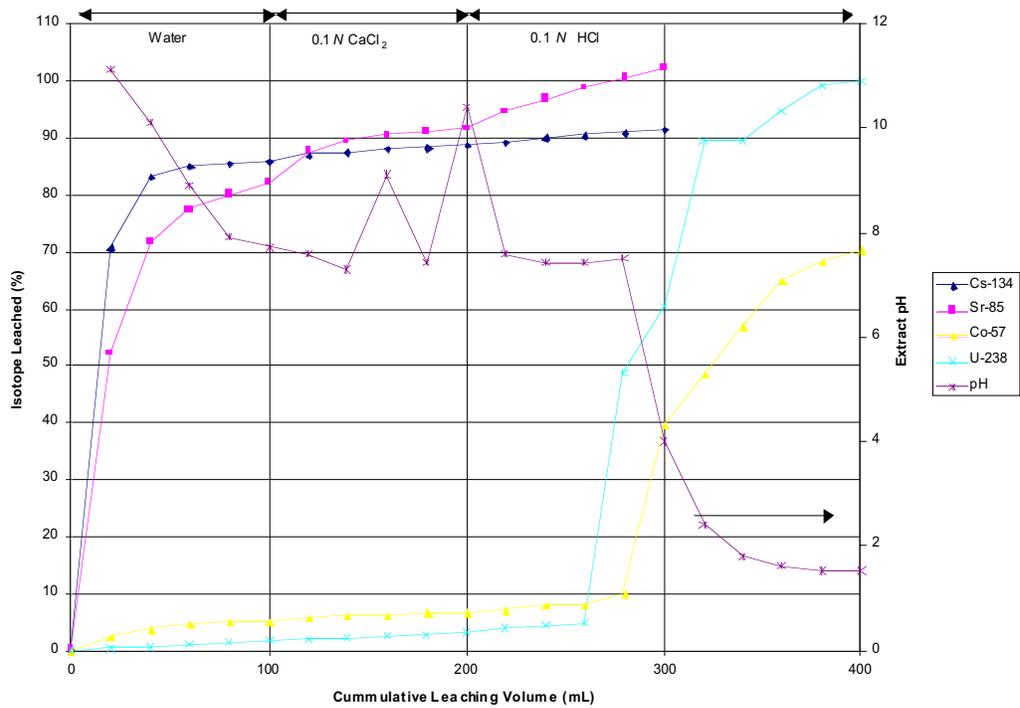


Figure 2. Sequential batch extraction of radioisotopes from Portland cement with water, dilute calcium chloride, and dilute hydrochloric acid.

the first five treatments with dilute HCl. At present, only this “baseline” of non-thermally altered cement extraction behavior has been completed. It is anticipated that significant changes in these extraction behaviors will be observed as cement is heated when significant mineral alterations are known to occur.

In the mechanics and thermodynamics subtask, the difficulty of modeling the heating of cement and concrete lies in the need to couple the heat and mass transport of pore water and various radionuclides it may contain. Previous studies in the field of nuclear reactor accidents used the finite element approach, which is plagued by instabilities at the sharp front of heating and propagating pressure. To overcome these difficulties, the finite volume method developed in the fields of geophysics and groundwater flow has been adopted and extended for the present coupled multiphase transport problem. By the time of this report a one-dimensional computer program has been developed, debugged, and successfully tested on example problems. The simulation of pore water is based directly on its known thermodynamic properties, with corrections for capillarity and adsorption. It is gratifying that the response converges well with volume refinements and is very stable.

At the same time, an alternative solution which has the potential of greater insight has been developed, utilizing the concept of moving layers with jumps in flux and pore water content at the interfaces. The velocity of the interfaces is obtained from Stefan type jump conditions. Analytical formulation of this second approach has been completed and numerical studies are starting.

Planned Activities

In the chemical subtask, the behavior of the four radionuclides, after thermal treatment of cement, will be followed through the sequential extraction procedures. Heating rates and additions of volatility-enhancing chemicals will be tested on additional cement samples. Addition of aggregates, both siliceous and carbonate types, on the volatility and extraction behavior of these radionuclides will also be examined. Concomitant measurements of cement compressive strength will be obtained to correlate with the observed chemical changes mediated by thermal treatment.

In the modeling subtask, the two computational approaches will be tested by simulating the existing test data on rapid heating with pore pressure measurements. The next phase will deal with the calculation of thermal and hydraulic stresses caused by rapid heating, with a view to analyze fracture propagation and spalling.

Information Access

No publications from this project have yet been completed. Information on chemical extraction and radionuclide modeling for soil and for cement (coming soon) can be obtained at <http://www.ornl.gov/LANGMUIR/bpsintro.htm>.